COMPLEXITY IN THE EARLY SOLAR SYSTEM AS RECORDED IN METEORITIC ORGANIC MATTER.

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Introduction: The organic matter contained within carbonaceous chondritic meteorites has the potential to provide a unique perspective of chemical processes that occurred early in Solar System history. Recently, for example it was concluded that the significant chemical structural variation across Insoluble Organic Matter (IOM) isolates from four petrologic type 1 and 2 chondrites recorded a history of differential low temperature chemical oxidation within the meteorite parent body [1]. The most oxidized IOM in this study was isolated from Tagish Lake, the least oxidized, hence most primitive IOM came from the CR2, EET92042. This study employed solid state ^{13}C Nuclear Magnetic Resonance (NMR) Spectroscopy, a very powerful tool for studying the chemistry of complex organic solids, but a technique that requires relatively large quantities of sample. In order to expand our studies to a much wider range of IOM obtained from a large suite of meteorites including CV, CO, Ordinary, and Enstatite Chondrites we have turned to using Carbon X-ray Absorption Near Edge Spectroscopy (C-XANES) utilizing a Scanning Transmission X-ray Microscope (Beamline 5.3.2) at the Advanced Light Source, Lawrence Berkeley National Laboratory. To date we have acquired C-, N-, and O-XANES on 23 different IOM isolates from all major chondritic groups, spanning a range in petrologic type from 1 to 4. C-XANES employing STXM has been used previously in the study of meteorite IOM and organics in IDP's by Flynn and colleagues [2, and references therein].

Experimental: Meteorite IOM was isolated from crushed meteorite via dissolution in concentrated CsF/HF solutions as described previously[3]. In order to perform C-, N-, and O-XANES with STXM, ultrathin sections on the order of 100 nm thick. Meteorite IOM is highly nanoporous in nature, therefore it is necessary to embed the IOM in sulfur as a support. Sectioning was performed using a Leica Ultracut microtome and a diamond knife. Sections were collected on SiO coated TEM grids. The sulfur was removed via sublimation prior to analysis. C-, N-, and O-XANES were performed at Beamline 5.3.2, a STXM with a bending magnet X-ray source [4].

C-XANES provides an assessment of functional group concentrations by virtue of the intensity of specific electronic transitions, e.g. the aromatic carbon 1s-π* transition [5].

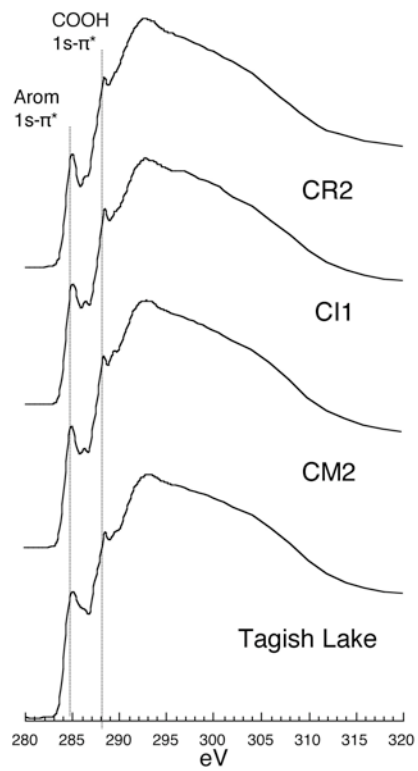

Figure 1: C-XANES spectra of IOM from EET92042 (CR2), Orgueil (CI1), Murchison (CM2), and Tagish lake (C2). Spectral bands associated with aromatic and carbonyl 1s-π* transitions are highlighted.

One parameter one would like to determine for a broad range of IOM is an estimate of the fraction of aromatic carbon. Both oxidative and thermal processes tend to lead to an increase in aromatic carbon relative other more labile functional groups. From previous NMR studies [1] the fraction of carbon in aromatic and/or olefinic functional groups is well determined; thus we can use these data to establish whether we can obtain a correlation between the intensity of the aromatic or olenfinic 1s–π* transition with the NMR derived aromaticities. C-XANES data for the four meteorite IOM samples reported on recently are shown in Figure 1. If we define the aromatic/olefinic carbon 1s-π* as spanning from 284 to 287.2 eV we can derived normalized (to the intensity at 340 eV) 1s-π* intensities. We observe in Figure 2 a reasonable correlation between the 1s-π* intensities and NMR derived aromaticity values.

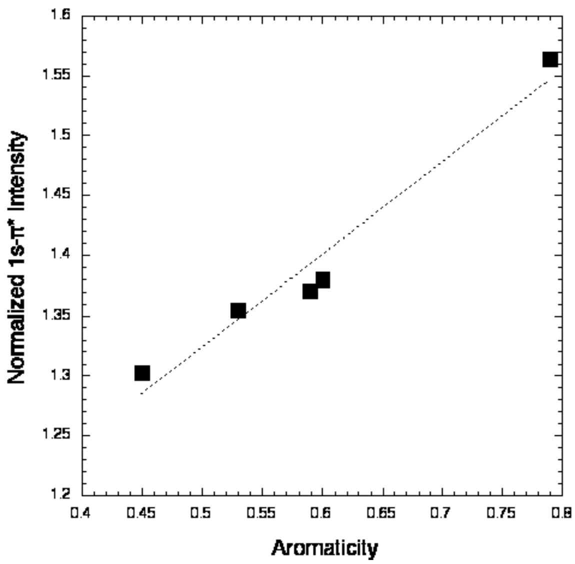

Figure 2. Correlation of NMR derived aromaticity with the intensity of C-XANES 1s-π* absorption for EET92042, Bells, Murchison, Orgueil, and Tagish Lake IOM.

In Figure 3 the normalized 1s-π* intensity are presented, sorted against IOM sample by intensity. As would be expected sample EET92042 a CR2 has the least intensity consistent with what we believe to be the highly primitive nature of the IOM in this meteorite. Joining EET92042 are Orgueil, Bells, Murchison, and GRO95577 (a CR1). All of the CV's, Ordinary Chondrites, and CO's exhibit greater 1s-π* intensity. Joining these are Tagish lake (an oxidized C2) and Y86720 (a thermally metamorphosed CM). The petrologic type 3 meteorites exhibit an enormous range in 1s-π* intensity. The relative ranking of type 3 IOM in terms of 1s-π* intensity is highly counter to what would be expected based on mineralogic indicators of meteorite alteration [e.g. 5]. For example, the intensity of the 1s-π* for the unprocessed CO ALH77307 is significantly more intense than that of the more processed CO ALH77003; The ordinary chondrite Bishunpur has a considerably more intense 1s-π* transition than Allende. These data suggest that the IOM records a chemical record that, at least in part, differs from that of the inorganic matrix assemblage. Of course the intensity of the 1s-π* is only part of the story, the C-XANEs spectra of these IOM contain considerably more chemical information, e.g. types and quantities of oxygen containing organic functional groups. These data coupled with N-, and O-XANES data, elemental chemistry, and isotopes leads to the conclusion that the chemical history recorded in IOM is highly complex although not intractable.

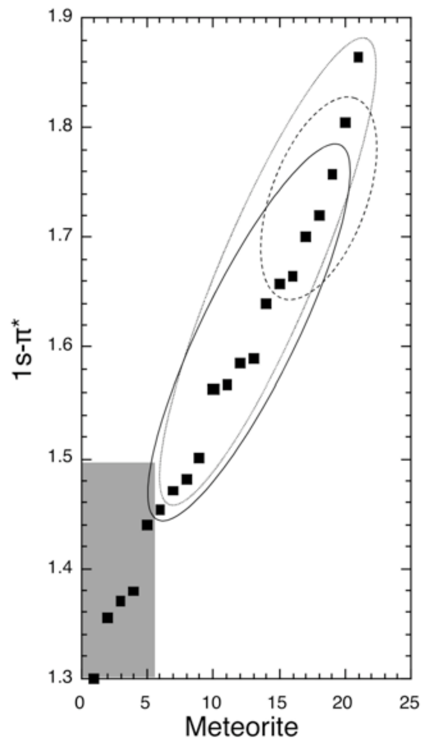

Figure 3: The intensity of the 1s-π* absorption for IOM from 21 different meteorite IOM's, sorted by intensity. The gray box groups the most primitive IOM, the solid line region encircles ordinary chondrite IOM, the fine dashed ellipse groups the CV IOM, and coarse dashed ellipse groups the CO IOM. The samples are: 1)EET92042, 2)Bells, 3) Murchison, 4) Orgueil, 5) GRO95577, 6) MET0489, 7) Kaba, 8) Y86720, 9) Allende, 10) Tagish Lake, 11) Vigarano, 12) Semarkona, 13) Mokoia, 14) Kainsaz, 15) ALH77003, 16) Bishunpur, 17) Chainpur, 18) Krymka, 19) Tieschitz, 20) AlH77307, 21) Leoville.

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